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CHARACTERIZATION BY XRD ANALYSIS, MOLECULAR VIBRATION AND ELECTRICAL ROPERTIES OF ALUMINIUM DOPED NICKEL-FERRITES PARTICLES

Ye Wint Htun[1], Sandar Oo[2], Soe Soe Han[3], Aye Aye Thant[4]

Abstract

Aluminium doped Nickel-ferrite nanoparticles of general formula: $NiAl_xFe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) have been synthesized by sol-gel auto combustion method involving final sintering at 900ºC with a heating rate of 5 hours. The phase formations of the sintered samples were characterized by X-ray Diffraction analysis. The X-ray diffraction patterns confirmed formation of ferrite with single phase cubic spinel structure with the appearance of small peaks representing secondary phases. The electrical properties of samples have been measured using Fluke- 189 LCR meter in the low frequency range (1-1000) kHz and higher frequency range (1 kHz to 10GHz).

Keywords: Ni-ferrite, sol-gel auto combustion technique, XRD, electrical properties.

Introduction

Ferrites particles have improved catalytic, dielectric and magnetic properties, as they possess high resistivity and negligible eddy current losses [Cullity, B.D. (1989)]. Magnetic particles promise some interesting applications, such as in high frequency devices, magnetic fluids, high density magnetic recording. In the present investigation we have employed sol-gel auto-combustion method to synthesize Al doped nickel ferrite nano-particles [Ghasemi, A., Ekhlasi, (2014)]. The sol-gel auto-combustion method is used to speed up the synthesis of complex materials. It is a simple process, which offers a significant saving in time and energy consumption over the traditional methods, and requires less sintering temperature. This method is employed to obtain improved powder characteristics, better homogeneity and narrow particle size distribution, thereby influencing structural, electrical, and magnetic properties of spinel ferrites [Auzans, E., Zins, D., Blums, E., & Massart, R. (1999)].

1. Assistant Lecture, Department of Physics, Ma U Bin University
2. Dr, Lecture, Department of Physics, University of Yangon
3. Dr, Lecture, Department of Physics, University of Yangon
4 Dr, Professor, Department of Physics, University of Yangon

In the inverse spinel structure of $NiFe_2O_4$ the tetrahedral sites are occupied by ferric ions and octahedral sites by ferric and nickel ions. The investigation of aluminum substituted nickel ferrite $NiAl_xFe_{2-x}O_4$ was not well documented and we present here the synthesis, structural characterization and also the basic magnetic properties of $NiAl_xFe_{2-x}O_4$ nanoparticles.

Experiment

Nano-crystalline powders of $NiAl_xFe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0) were prepared by sol-gel auto-ignition method. The Citric acid ($C_6H_8O_7$. H_2O), Nickel Nitrate ($Ni(NO_3)_2.6H_2O$), Ferric Nitrate ($Fe(NO_3)_3.9H_2O$), Aluminium Nitrate ($Al(NO_3)_3$. $9H_2O$). Nano-crystalline powders of $NiAl_x$ $Fe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) have been prepared by have been used as starting materials. The molar ratio of metal nitrates to citric acid has been taken as 1:3. The metal nitrates have been dissolved together in 100 ml of de-ionized water to get a clear solution. An aqueous solution of citric acid has been mixed with metal nitrates solution, then ammonia hydroxide solution has been slowly added to adjust the pH at 7. The mixed solution has been moved on to a hot plate with continuous stirring at 90ºC - 100ºC. During evaporation, the solution became viscous and finally formed a very viscous brown gel. When finally all remaining water was released from the mixture, the sticky mass began to bubble. After several minutes the gel automatically ignited and burnt with glowing flints. The decomposition reaction would not stop before the whole citrate complex was consumed. The auto ignition was completed within a minute, yielding the brown-colored ashes termed as a precursor. The as-prepared powder has been annealed at 700ºC for 3 hrs to get single phase spinel product and used for further characterization. Then, the powder has been pressed into pellets by hydraulic press at a pressure of 5 tons. The final sintering has been performed at 900°C for 5hrs.The phase formations of the sintered samples have been characterized by X-ray Diffraction (XRD) analysis and the electrical properties.

Figure 1: The starting solutions in the preparation of Aluminium doped Nickel-ferrite particles by sol-gel auto-combustion method

Figure 2: The starting solutions in the preparation of Aluminium doped Nickel-ferrite particles by sol-gel auto-combustion method

Results and Discussion

Phase formation

Aluminium doped Nickel-ferrite nanoparticles, $NiAl_xFe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) have been synthesized by sol-gel auto combustion method and characterized using X-ray diffraction (XRD). X-ray diffractograms of $NiAl_xFe_{2-x}O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) samples. This XRD diffractograms display formation of well nano-crystalline undoped and alumina doped $NiFe_2O_4$ to be single cubic spinel phase with reflection planes of (2 2 0), (3 1 1), (4 0 0), (4 2 2), (5 1 1) and (4 4 0). The XRD patterns clearly indicate that the prepared samples contain cubic spinel structure only. The strongest reflection peak has resulted from the (3 1 1) plane that indicates the spinel phase. An increase of the aluminium content resulted in a measurable decrease in the degree of crystallinity of spinel nickel ferrite phase with subsequent a decrease in the intensity of its diffraction peaks. Some diffraction planes such as (2 2 0) and (4 4 0) planes are more sensitive to the cations distribution on tetrahedral and octahedral sites, respectively. [10] The sizes of crystallites in the sample have been evaluated by using the FWHM of the most intense peak (220) (311) (400) (511) (440). The results are shown in Table 1. Further, it is observed that particle size decreases with an increase in nonmagnetic Al substitution.

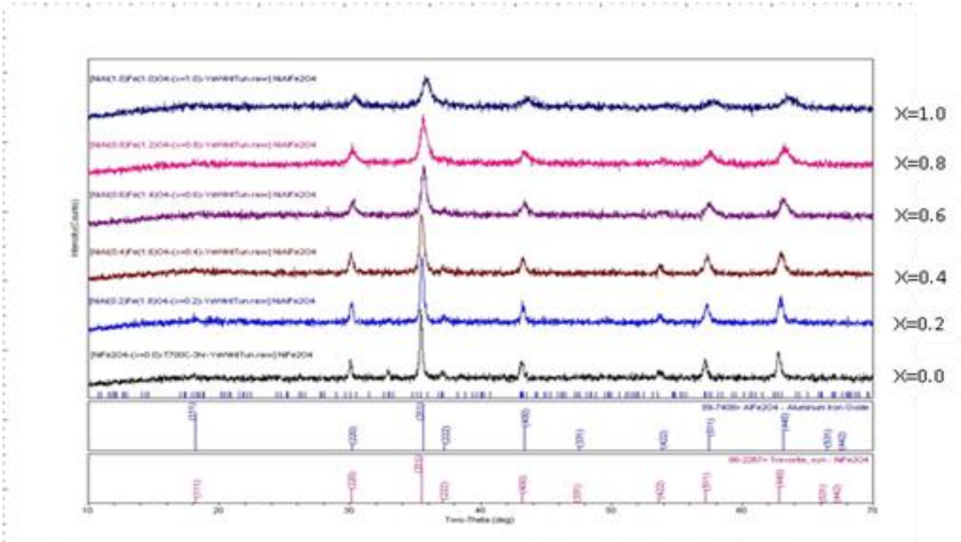

Figure 3: XRD spectra for of $NiAl_xFe_{2-x}O_4$ particles at 900°C for 5 hours (The inset shows the peak-shift in 311plane.)

Table 1: Comparison of crystallite size for different compositions

Composition (x)	Lattice parameter (a=b=c) Å	Crystallize size D (nm)
0.0	8.31	57.05
0.2	8.29	46.07
0.4	8.28	54.68
0.6	8.23	34.58
0.8	8.20	31.26
1.0	8.19	30.1

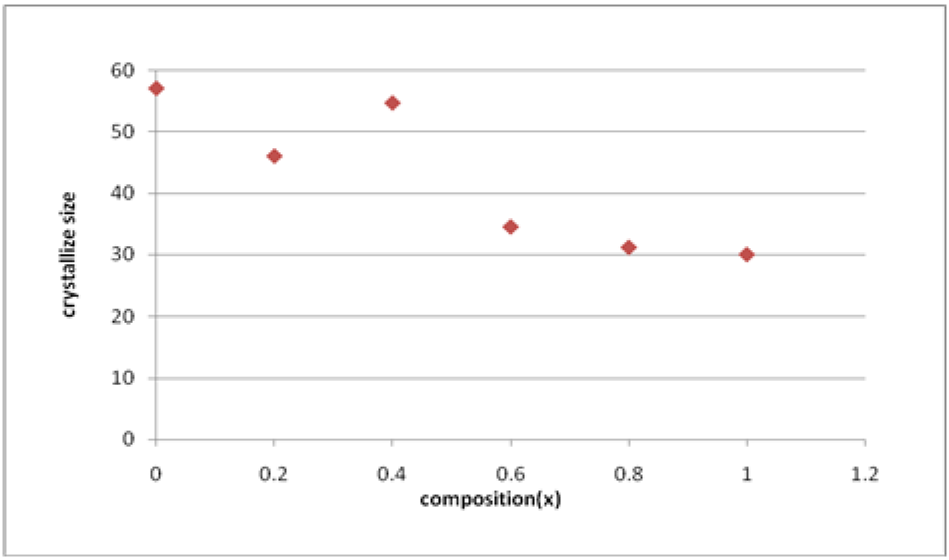

Figure 4: Variation of Crystallite size with chemical composition x

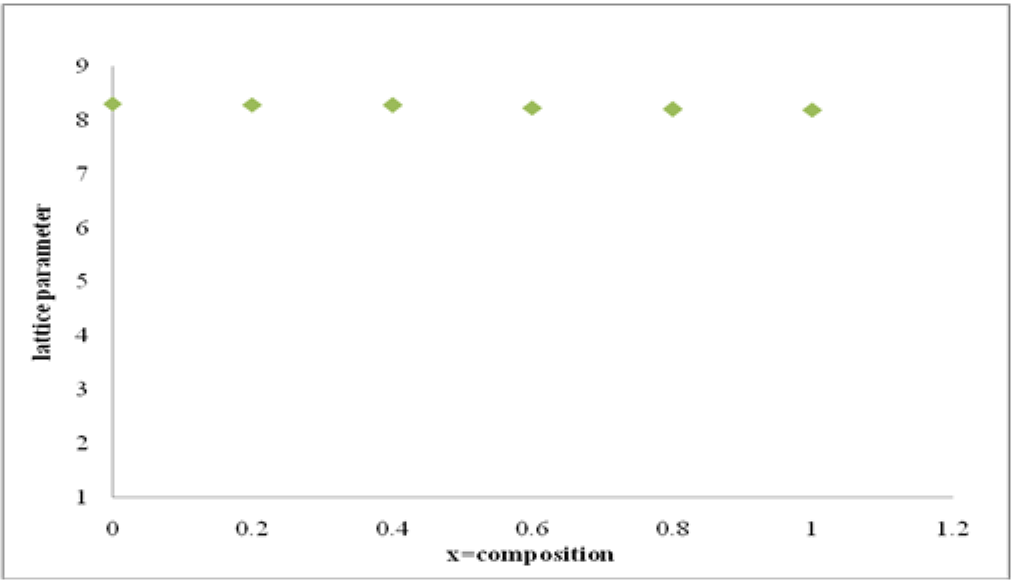

Figure 5: Variation of lattice parameter with chemical composition x

Study on Frequency Dependent Resistivity

Aluminium doped in Nickel ferrite nano-particles have also been synthesized by sol-gel auto combustion method. The measurements on electrical resistance and capacitance were performed on the final sintered pellet at 900□ C. The resistances were measured using Fluke- 189 LCR meter. The resistivity of each sample was calculated by measuring the dimensions and resistance of the sample. Firstly, the cross sectional area (A) and the thickness(t) of each sample were measured accurately. The resistivity of each sample was calculated by using the following relation:

$$\rho = \frac{RA}{t} \tag{1}$$

where R is the resistance of the sample, A is the area of the electrode in contact with the sample (πr^2) and t is the thickness of the sample.

The conductivity σ of the sample ($NiAl_xFe_{2-x}O_4$) was determined from the measure resistance value and sample dimension by using the relation:

$$\sigma = \frac{1}{\rho} \tag{2}$$

The frequency dependent DC resistivity (the inverse of conductivity) and dielectric constant for all sample which are final-sintered at 900°C at lower frequency range 1kHz-1000kHz (1kHz-1MHz) and 1kHz to 10GHz. These grain boundaries are more active at lower frequencies; hence, the hopping frequency of electrons between Fe^{3+} and Fe^{2+} ions have less at lower frequencies. [6] The resistivity shows fluctuation in the lower frequency region and there is almost no variation in the higher frequency region. This is normal behavior of ferrite as grain boundary hinders the conduction mechanism and this effect disappears in the higher frequency region.

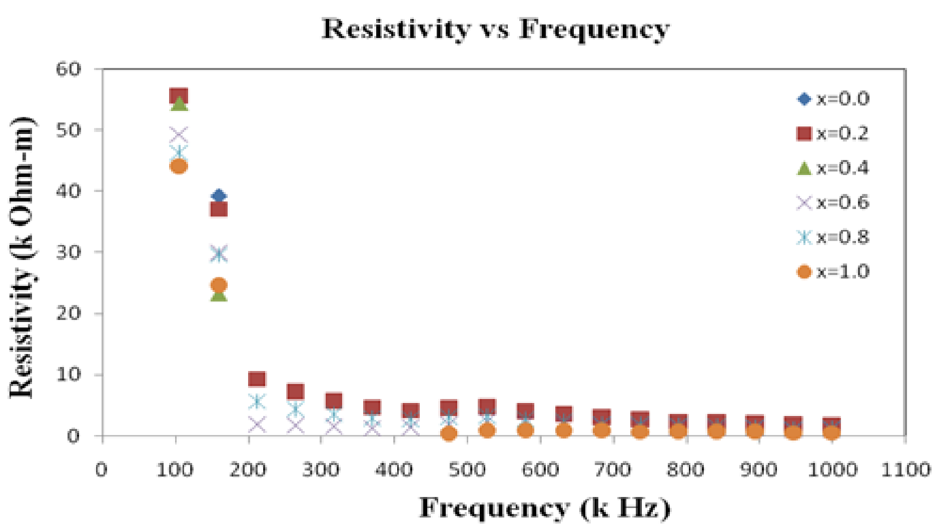

Figure 6: The variation of resistivity with lower frequency range (1kHz to 1000kHz) for (x=0.0 to 1.0)

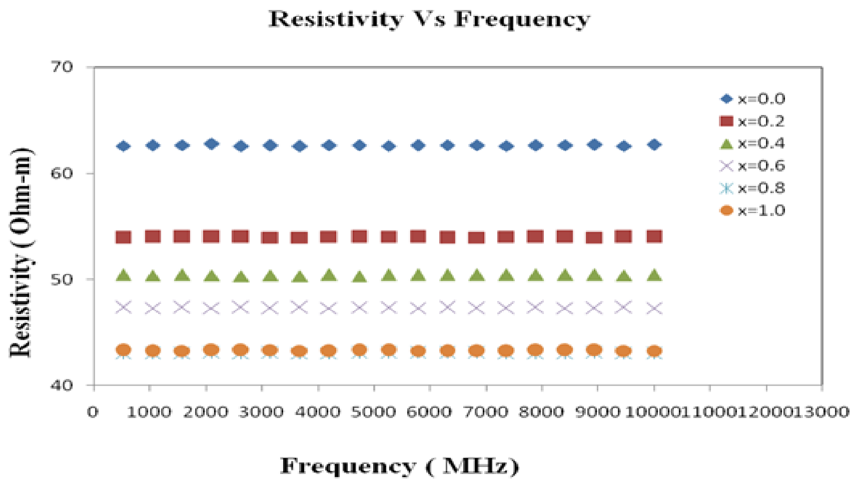

Figure 7: The variation of resistivity with higher frequency range (1kHz to 10GHz) for (x=0.0 to 1.0)

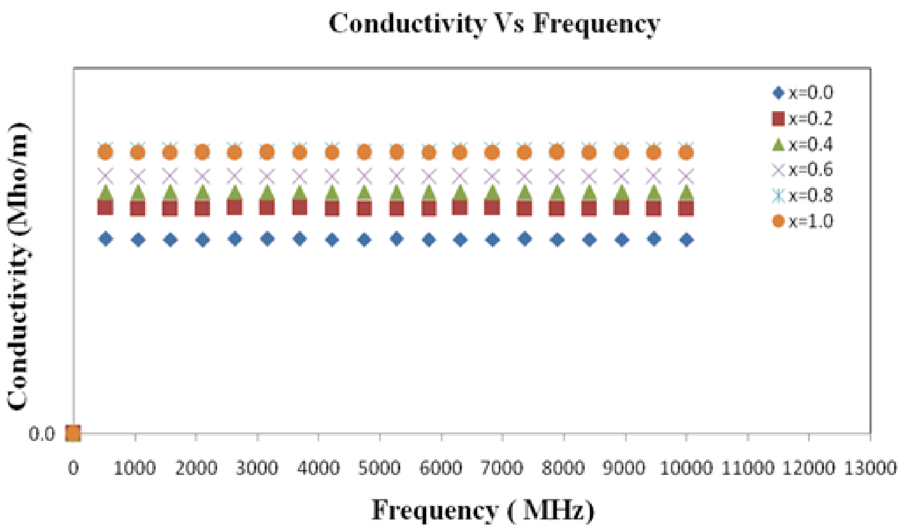

Figure 8: The variation of Conductivity with higher frequency range (1kHz to 10GHz) for (x=0.0 to 1.0)

Conclusion

The XRD patterns clearly indicate that the prepared samples contain cubic spinel structure only. The obtained average crystallite sizes decreases with increase in nonmagnetic Al substitution. An increase of the aluminium content resulted in a measurable decrease in the degree of crystallinity of spinel nickel ferrite phase with subsequent decrease in the intensity of its diffraction peaks. The resistivity shows fluctuation in the lower frequency region and there is almost no variation in the higher frequency region. This is normal behavior of ceramics as grain boundary hinders the conduction mechanism and this effect disappears in the higher frequency region.

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References

Auzans, E., Zins, D., Blums, E., & Massart, R. (1999). Synthesis and Properties of Ferrite. *Journal of Materials Science,* 34(6), 1253-1260.

Cullity, B.D. (1989). *"Elements of X-ray Diffraction*. Addison-Wesly Publishing Co. Inc.

Ghasemi, A., Ekhlasi, S. and Mousavinia, M. (2014). Structural and Magnetic Properties of Nickel Ferrite Synthesized by Using the sol-gel Auto-Combustion Method. *Journal of Magnetism and Magnetic Materials*, 354, 136-145.

H.P.Klug, L.E.Alexander, 1997, X-Ray Diffraction procedures for Polycrystalline and Amorphous materials, Wiley, New York, NY, **P.637**.

J.Smit & H.P.J.Wijn, (1959). "Ferrites" Physical Properties of Ferrimagnetic Oxides in

KOOPS C.G., Phys. Rev. Lett., 83 (1951), 121.

Mathew, G., Asha, M.J., S.Nair, Joy, P.A., Anantharaman, M.R. & Magn.Magn.Mater. J. (2006). 302**,** 190.

Niu, Z. P., Wang, Y. & Li, F. S. (2006). Magnetic Properties of Nano Crystalline Co-Ni Ferrite. *Journal of Materials Science*, 41(17), 5726-5730.